THE GEMMOLOGIC QUALIFICATION OF SOME VARIETIES OF PEGMATITE AND HYDROTHERMAL MINERALS IN PORTUGAL

Leal Gomes, C., Dias, P.

Universidade do Minho, CIG-R, Escola de Ciências, Gualtar, 4710-057 BRAGA, Portugal

Keywords: pegmatite, *subsolidus* gem stage, pseudomorphic recrystalization, gem grade

INTRODUCTION

In northern Portugal several pegmatite bodies and their related hydrothermal veins and metasomatic paragenesis, contain gem varieties of beryl, tourmaline, quartz, feldspar, phosphates and oxides. Most mineral deposits that include gemstones are genetically related to the evolution of different types of Variscan granites (fig. 1). In this study some ornamental varieties of feldspars, Li minerals (mainly tourmaline) and Be-minerals (especially beryl), were studied in detail by X-ray diffraction and chemical analysis both in separated phase mode and electron microprobe. Gemmy qualification was attributed through carving and lapidary assay.

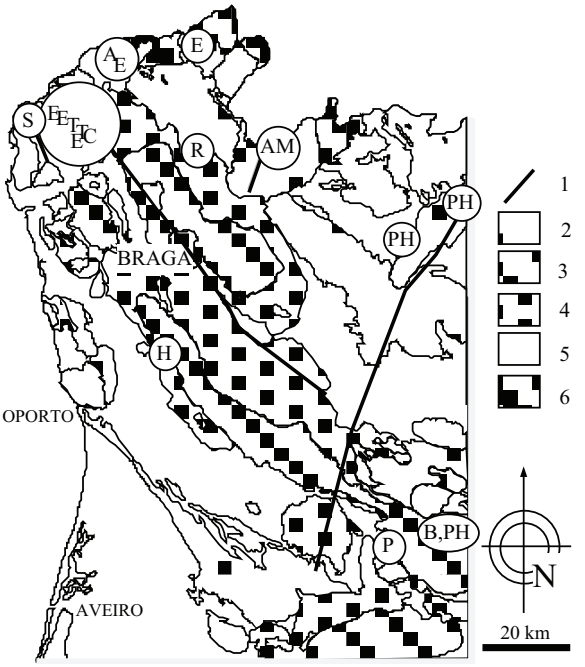

Figura 1 – Geographic and geological distribution of the most important occurrences of gemstones: E – emerald; S – sapphire; B – noble beryl; AM – aqua marine; PH – phenakite; C – chrysoberyl; T – coloured tourmaline; P – petalite; A – amazonite; H – hyaline quartz and inclusion quartz; R – pink quartz. Symbols: 1- major tectonic lineaments; 2 – post- tectonic granites relatively to D_3 Variscan folding phase; 3 - D_3 late-tectonic granitoids related with ductile shear; 4 - D_3 syntectonic granitoids related to ductile shear; 5 – syntectonic two-mica granites related to D_3; 6 – restitic two-mica granites and migmatites

AMAZONITE

The amazonitic colour (bluish green to green) of some potash feldspars typical of miarolitic pegmatites is attributed to petrogenetic specialization of parent D_3 post-tectonic granites. The emplacement of amazonitic pegmatites may occur inside or outside granite outcrops and is generally related to the relaxation and cupola collapse of plugs and stocks of post-tectonic emplacement. Some light coloured amazonites show X-ray monoclinic diffraction patterns and disordered structure. The others are triclinic and ordered, regardless of their colour and position in the internal structure of the pegmatite. Deuteric-superimposed feldspar reddening can completely obliterate the standard green colour. Green-blue highly ordered amazonite with Pb over 350 ppm (Table 1) have the amazonitic colour due to dimeric complexes such as $[Pb\text{-}Pb]^{3+}$. The amazonites showing Pb content lower than 350 ppm and high H_2O content (high LOI) are less ordered and yellow. The green colour can result of structural coupling of H_2O and Pb ($2K^{+} => Pb^{2+} + H_2O$). Reddening may be related to reheat (diffusion of Pb) and leaching of microcline.

There is a negative correlation between Pb and Rb or Cs. Considering that Rb and Cs tend to increase with fractionation then the amazonitic colour (interpreted as the result of an amazonitization process) is primitive and may occur immediately after the formation of more peripheric and earlier units of these pegmatites.

TOURMALINE

There are consistent relations between colour, gemmological quality and composition of coloured tourmalines from Portuguese pegmatites. Varying Li contents provide a discrimination of gemmological types and qualifications. Gemmy varieties are elbaite, F-elbaite, liddicoatite and rossmanite, Some olenites may also achieve the gem grade. As seen in fig. 2A the most clear and transparent tourmalines are the richest in Li (Fig. 2A). The Li/Fe ratios of primary dark green-grey, Na rich "urubu" tourmalines mark the transition between good gem quality tourmalines from well fractionated LCT paragenesis and tourmalines of lower quality which are from earlier paragenesis rich in microcline. The liddicoatite trend (Fig. 2) is only for pink tourmalines coexisting with cookeite and/or lepidolite in late replacement units (re-deposited, re-crystallized or pseudomorphic). The Li and Ca rich tourmalines are the lightest and pinkest. Fluctuations in quality within each colour type are related to varying contents of F and Al (Fig, 2C).

Table 1 – Selected compositions of some gemstones: Am – amazonite; Tou – tourmaline; Em – emerald; Ber – noble beryl; Chry – chrysoberyl. Notes: vst – vestigial, oxides in wt%; elements in ppm; amazonite was analysed in separated phase hand picked; all other minerals analysed by electron microprobe; *- calculated by stecheometry; all Fe as Fe_2O_3.

	Am	Tou 1	Tou 2	Tou 3	Tou 4	Em 1	Em 2	Em 3	Em 4	Ber 1	Ber 2	Ber 3	Chry
SiO_2	65.96	39.58	37.17	36.51	39.17	65.67	66.45	66.97	65.91	67.79	67.35	66.90	-
Al_2O_3	17.67	42.15	37.4	35.91	42.86	14.86	15.66	16.77	14.77	18.27	18.59	18.69	82.10
MgO	0.00	0.04	0.06	0.28	0.05	2.40	1.83	0.92	1.73	0.05	0.05	0.07	0.08
CaO	0.00	0.18	0.10	0.00	0.18	0.15		0.01	0.02	0.00	0.00	0.00	-
Na_2O	2.28	1.73	2.45	1.31	1.58	1.43	1.34	0.90	1.32	0.23	0.24	0.29	-
K_2O	12.56	0.02	0.00	0.03	0.03	0.28	0.11	0.08	0.09	0.02	0.01	0.03	-
TiO_2	0.00	0.01	0.02	0.04	0.00	-	-	-	-	-	-	-	0.05
FeO	-	0.07	5,19	13.01	0,14	0.60	0.37	0.56	1.60	0.23	0.19	0.33	0.30
Cr_2O_3	-	-	-	-	-	0.14	0.08	0.16	0.15	0.00	0.00	0.00	0.02
Sc_2O_3	-	-	-	-	-	-	0.05	0.07	0.01	0.00	0.02	0.00	-
V_2O_3	-	-	-	-	-	0.05	0.03	0.16	0.02	0.00	0.00	0.00	-
Cs_2O	-	-	-	-	-	0.23	0.13	0.10	0.31	0.02	0.09	0.14	-
F	-	0.89	1.09	0.23	0.65	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
H_2O*	n.d.	3.50	3.18	3.56	3.61	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
B_2O_3*	n.d.	11.37	10.70	10.64	11.36	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Li_2O*	n.d.	2.25	1.71	0,42	2.03	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
BeO*	n.d.	n.d.	n.d.	n.d.	n.d.	13.63	13.76	13.89	13.61	14.02	13.99	13.95	-
Total	98.39	101.80	99.08	101.94	101.66	99.43	99.81	100.59	99.54	100.63	100.53	100.41	82.64

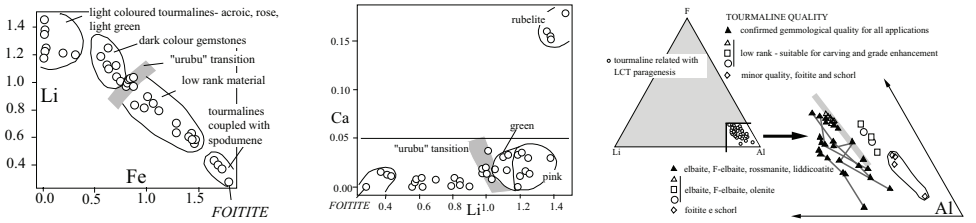

Figure 2 – Compositional trends (in apfu) observed in gem tourmalines from LCT aplite-pegmatites from the pegmatite fields of Minho and Beira Provinces.

Studies of fluid inclusions (FI) in tourmalines from Serra de Arga (LCT pegmatites) revealed primary aquo-carbonic inclusions with not negligible contents of CH_4 (salinity of 3.5% weight eq. NaCl) and aqueous inclusions with salinity from 2 to 3% weight eq. NaCl (Tf_{ice} = -1.2° to -2.0°C). Thermometry suggests temperatures of crystallization of green elbaite between 260° and 300°C. Aquocarbonic inclusions indicate pressures in the range 850-1300 b for more evolved tourmaline. The primitive nodular blue aggregates have inclusions with the highest homogenization temperatures, in the range 330°C (periphery) - 360 °C (core) and pressure > 1500 b. Several differences in the thermal regime are recorded in FI: elbaite stretching – 285º to 296ºC; secondary inclusions in olenite – 240º to 270°C; epitaxic overgrowth of pink elbaite on green elbaite – 249º to 262° C; corrosion/redeposition of hydrothermal acroic elbaite to rossmanite – 181º to 209º C; precipitation of pink liddicoatite – 172º to 270° C; recrystallization of green elbaite - t < 200 ° C.

PETALITE

Puga et al (2003) suggested the gemmological interest of petalite with "cats-eye" effect from Queiriga pegmatite (Vila Nova de Paiva). In fact, the incipient transformation of petalite into fibrous spodumene during the isochemical reaction (subsolvus to subsolidus) petalite⇔ spodumene + 2 quartz (in response to pressure and temperature changes during 3rd Variscan deformation phase), produces layers and bands with transverse fibres generating a satin gloss in crenulation related to kink bands.

EMERALD

In Northern Portugal the occurrence of emerald in leucocratic veins and metasomatic masses is located in shear corridors affecting metasedimentary and metavolcanic Silurian terranes. Porphyroblastic emeralds have phlogopite, biotite and zircon inclusions and particularly spinel exsolutions with Cr, Fe, Al and Zn (including chromite). Emeralds that occur in skarn deposits (segregated plagioclase veins) have apatite, zircon and oligoclase inclusions besides phlogopite. The compositions of emerald from porfiroblasts and veins linked to meta-ultramafic lithologies are clearly the most magnesian and homogeneous in composition (table 1). Fe richest crystals occur close to volcanogenic protolith with disseminated sulphides. Possibly the MgO-FeO path distinguishes protoliths and processes of emerald formation. Porphyroblasts have the highest and the most dispersed values of V and Sc.

OTHER BERYL WITH GEM QUALITY

All gem beryl examined present a depletion in the contents of some trace elements such as Fe and Cs if these are compared to the contents of non gemmological or primary beryl (Fig. 3). Therefore, in general the acquisition of gemmological quality is related to a decrease in the total of trace elements, although aquamarine fractionation (Gerês pegmatites) always retains a slightly higher content of Fe, as appropriate chromophore for deep blue. The heliodore (yellow to honey coloured) from Senhora de Assunção (Satão) beryl is not compositionally

distinguishable from pale blue beryl from the same deposit. Tests of flame heating change the yellow to primitive blue of primary beryl. Electron microprobe analysis shows similar compositions when yellow primitive phases and blue heated phases are compared. It seems reasonable to assume that the yellow colour comes from an increase in the volatile content of beryl and/or a change of Fe oxidation state. This combination of events could be facilitated by the structural disorder caused by radioactive bombardment from contiguous U and Th bearing phases.

Also in Senhora de Assunção some of the clearest peaces are of 2nd generation and result from the re-mobilization/ pseudomorphic re-deposition of primary giant beryls in favourable structural conditions. Quality maintenance depends on effective sealing of the hydrothermal cells after the acquisition of gem grade. In Leal Gomes (2003), the study of FI of primary giant-beryls, and late hydrothermal beryl, phenakite or bertrandite, shows frequency peaks of homogenization temperatures between 380°C (primary beryl - marginal zone) and 180ºC (miarolitic goshenite from the quartz core). According to paragenetic equilibrium, the presence of bazzite and thortveitite mark the highest temperature of stability of "industrial" beryl of primary marginal zone (~ 500ºC). The lowest temperature of arrest of primary FI (~ 270ºC) – coincides with beryl destabilization by acidic solutions following the reaction, *4 beryl + 7H_2O ⇔ 3 bertrandite + 4 pyrofilite(or equivalent) + 2 QZ*. So, 280 to 290°C, are reference temperatures for the re-crystallization of hydrothermal paragenesis, which include phenakite, euclase, bertrandite and gem beryl.

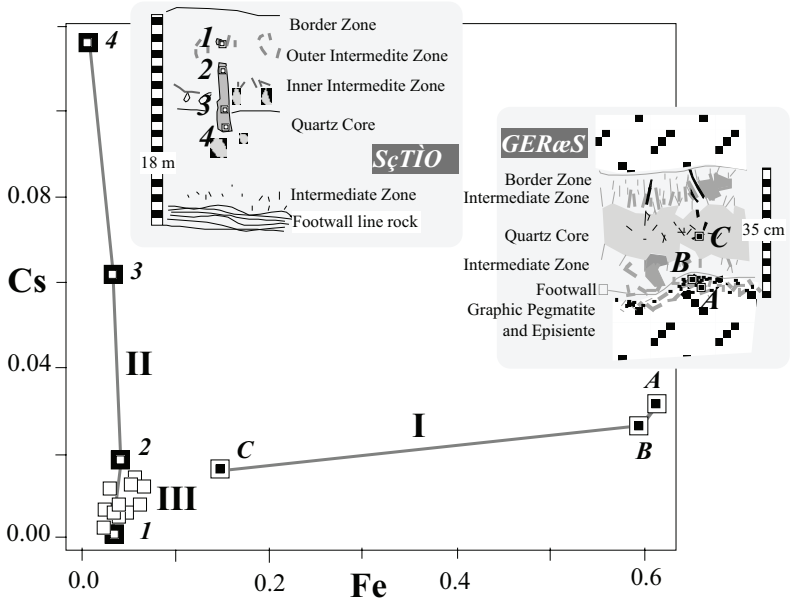

Figure 3 – Cs and Fe (apfu) diagram for beryl of different paragenetic positions and different types of pegmatites. I = A-B-C - trend for "aquamarine" in the quartz core of NYF pegmatites (Gerês) II = 1-2-3-4 - representative trend of primary beryl from Senhora de Assunção pegmatite. III – light beryl, which result from remobilization of primary giant beryl crystals and re-deposition in corrosion and miarolitic cavities. Note: in both cases host-rock is granite.

CHRYSOBERYL

Rare chrysoberyl occurs in association with porphyroblastic emerald and phenakite, presenting occasional green-red alexandrite effect. In these cases microprobe analysis showed traces of chromium content possibly related to contamination from metamorphic basic rocks that occur nearby.

CONCLUSIONS

In general, gemstone genesis depends on fractionation, mainly when hydrothermal, metamorphic or deformational phenomena affect primary pegmatitic paragenesis. The more interesting crystals have paler colours and simpler compositions. FI studies of pegmatitic deposits with gem beryl and tourmaline suggest that the acquisition of ornamental quality in most cases may have occurred at low temperature hydrothermal conditions (<280°C). In cavities the gemmological quality persists. Mainly the inclusion in clusters of sheet silicates preserve the quality of the crystals due to the dissipation of tensions trough the chaotic orientation of (001) cleavages and by constriction mitigation as a result of sheet silicates elasticity. There are cases, however, in which corrosion removes imperfections and deformations improving the gem grade. The clearest stones result from pseudomorphic secondary hydrothermal equilibrium of the primary unstable phases.

REFERENCES

Leal Gomes, C. 2003. Equilíbrio e evolução dos berilos do jazigo pegmatítico de Senhora de Assunção – Viseu. *Resumos, IV Congresso Ibérico De Geoquímica, Coimbra,* 47 – 49.

Puga, M. S., Leal Gomes, C., Vide, R. 2003. Modo de ocorrência e ensaios de aplicação industrial da petalite do jazigo pegmatítico da Queiriga – Sátão (Viseu). *Resumos, IV Congresso Ibérico de Geoquímica, Coimbra,* 196-198.